

UCN transport simulation in solid deuterium crystals

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Abstract

The extraction efficiency of ultracold neutrons from cryogenic moderators depends critically on the neutron transparency of the moderator material. A Monte Carlo simulation of the probability of the UCN going out from non-ideal (disordered) solid deuterium crystals has been performed. It was based on the use of a correlation function describing the density fluctuations in a disordered material, the latter being inferred from the measured very low neutron energy total cross sections for this material.

Ultracold neutrons [1] (UCN, the energy below $\sim 0.3\mu\text{eV}$), which can be contained in closed volumes, have found important applications in the investigation of fundamental properties of the neutron [2].

The best UCN source – ILL UCN turbine [3] yields maximum UCN density in a trap of $\sim 50 \text{ n/cm}^3$ and, respectively, the flux $\sim 5 \times 10^3 \text{ n cm}^{-2}\text{s}^{-1}$.

Recent years brought hope for significant progress in the intensity of very low energy neutron beams. This is connected with the possible use of the most effective cold moderators: solid deuterium or solid deuterocarbons at low temperatures of $\sim 5 - 10 \text{ K}$ for production of very cold neutrons.

The most promising and popular material for the UCN converter is solid deuterium. The first test of the UCN production in solid deuterium has been performed by the PNPI group [4], which then has been studying experimentally the UCN production in deuterium during many years [5-11].

The first estimate of the UCN production and up-scattering cross sections in solid deuterium was made in [12]. The phonon frequency spectrum of solid deuterium was calculated in [13] using the dispersion relations measured in [14]. Later calculations in the incoherent approximation [15] of the UCN scattering on phonons in solid deuterium reproduced the results of [13], this publication also

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contains the calculation of the UCN spin-flip up-scattering on para-deuterium with para-ortho transitions of deuterium molecules. The incoherent approximation and the density of states of [13] was used in the calculations of the UCN production in [16].

It was proposed that because of comparatively short neutron lifetime in solid deuterium (140 ms in pure ortho-deuterium at $T=0$ K) solid deuterium can find better application in a pulsed mode of the UCN production [17,9].

A solid deuterium UCN source was also demonstrated in the experiments in LANL [18], it is under construction at FRM-2 [19] and is in the stage of completion in PSI [20].

The incoherent model of neutron interaction in solid deuterium was generally confirmed in the later works [18, 21, 22, 23, 24].

Recently a re-evaluation of the neutron scattering in solid deuterium was performed in [25], it was based on the calculations in the frame of a rigorous model of coherent neutron scattering by deuterium nuclei. The resulting scattering cross sections obtained in this work are significantly different from the previous predictions [12, 13, 15] based on the incoherent approximation. Especially large difference between the incoherent approach and coherent one was found in the case of UCN up-scattering with phonon annihilation in the temperature range of 5-10 K: the first one 3-4 times surpassed the second one. These new calculations found some experimental confirmation in [26].

It was clear from the very beginning that structural homogeneity of a deuterium crystal is very important for the UCN yield from the UCN converter. The neutron scattering on structural inhomogeneities increases the time neutrons spend inside the UCN converter before leaving it, increasing the probability of their capture and up-scattering. It was discovered in [27] that the transmission of very low energy neutrons through solid deuterium depends critically on the way of preparation of a solid deuterium sample. The light transmission investigations of solid deuterium crystals were performed in [28]. It was demonstrated that optical quality of crystals depends on the speed of crystal growing - more transparent crystals were obtained when they were carefully grown over a time period of about 12 hours at temperature close to the triple point.

The total cross section in the UCN energy range for solid deuterium frozen from liquid state was measured in [29]. These measurements showed that the best transmission have the solid deuterium samples prepared in the process of very slow cooling - less than 1 K per hour. This procedure of obtaining good quality deuterium crystals was based on preliminary investigations described in [28]. Temperature cycling between 5 K and 10 K, and between 5 K and 18 K seriously

deteriorated the neutron transmission. It is obvious that crystal imperfections introduced by thermal cycling increased the UCN scattering. In contrast to this experience, the solid deuterium crystals used as UCN converters and condensed directly from gas phase significantly increased the UCN yield being subjected to repeated thermal cycling [22].

We consider solid deuterium crystals frozen in the process of very slow cooling in [29] as the most perfect of all possible samples prepared from liquid, and use the energy dependence of the total cross section (Fig. 2 of [29]) to infer the parameters characterizing structural inhomogeneity of the sample. These parameters are used further to simulate the probability for the UCN generated in a similar solid deuterium UCN source, to go out from the source to vacuum.

To obtain the UCN scattering cross section on structural inhomogeneities the experimental points of Fig. 2 of Ref. [29] were corrected for the incoherent elastic scattering on deuterium (2.05 b), the UCN capture by deuterium ($1.19 \text{ b}/v$) and hydrogen ($733 \text{ b}/v$) nuclei, the UCN up-scattering in the result of one-phonon annihilation ($0.83 \text{ b}/v$), and the spin-flip up-scattering on para-deuterium ($123 \text{ b}/v$), where v is the neutron velocity in medium in m/s. The cross sections of the latter processes were taken from [15]. All the microscopic cross sections here and further are given per one atom of respective element. The concentrations of hydrogen (0.05%) and para-fraction (1.4%) were taken as they were done in [29]. The atomic density of solid deuterium at 5 K was taken to be $6 \times 10^{22} \text{ cm}^{-3}$. The resulting neutron wavelength dependence of the UCN scattering on inhomogeneities is shown in Fig. 1.

The transmission of very slow neutrons through an inhomogeneous medium was studied previously by A. Steyerl [30]. It was demonstrated that transmission as a function of neutron wave length may be used to deduce characteristic parameters of inhomogeneities, in particular their size and density.

It is known that in the Born approximation the differential macroscopic cross section of elastic scattering for neutrons transmitting through an isotropic inhomogeneous medium has the form [31]:

$$\frac{d\Sigma_{el}}{d\Omega} = \frac{1}{\pi} \left(\frac{m}{\hbar^2} \right)^2 \int_0^\infty G(\rho) \frac{\sin(q\rho)}{q\rho} \rho^2 d\rho, \quad (1)$$

where m is the neutron mass, $q=|\vec{k}' - \vec{k}|$ is the neutron wave vector change, and $G(\vec{r}, \vec{r}') = \langle \delta U(\vec{r}) \delta U(\vec{r}') \rangle$, ($\rho = |\vec{r}' - \vec{r}|$) is the correlation function of fluctuations of the local neutron-medium interaction potential.

The latter is

$$U = \frac{\hbar^2}{2m} \sum_i 4\pi N_i b_i, \quad (2)$$

where N_i is the atomic density and b_i is the coherent scattering lengths of nuclei of the medium, so that $\delta U(\vec{r}) = U(\vec{r}) - \langle U(\vec{r}) \rangle$.

The total cross section after integration over solid angle with the account of the solid angle of the neutron detector is:

$$\Sigma_{el}(k) = 2 \left(\frac{m}{\hbar^2} \right)^2 \frac{1}{k^2} \int_0^\infty G(\rho) [\cos(2k\rho \cdot \sin\theta_0) - \cos(2k\rho)] d\rho, \quad (3)$$

where $2\theta_0$ is the angle of the neutron detector from the sample.

For the exponential correlation function

$$G(\rho) = G_0 e^{-\rho/\rho_0}, \quad (4)$$

where ρ_0 is the correlation length, we have the expression

$$\Sigma_{el}(k) = 2 \left(\frac{m}{\hbar^2} \right)^2 \frac{G_0 \rho_0}{k^2} \left[\frac{1}{1 + 4k^2 \rho_0^2 \sin^2 \theta_0} - \frac{1}{1 + 4k^2 \rho_0^2} \right] \quad (5)$$

for the total cross section, and

$$\frac{d\Sigma_{el}}{dq} = \frac{4}{k^2} \left(\frac{m}{\hbar^2} \right)^2 \frac{G_0 q \rho_0^3}{1 + q^2 \rho_0^2} \quad (6)$$

for the differential cross section.

This formalism was used for the interpretation of the measurements of the total cross sections for solid deuterium at 5 K [29]. The parameters of the correlation function of Eqs. (4) and (5) were inferred by the least squared method from the corrected experimental points for the macroscopic cross section of the UCN scattering on inhomogeneities of Fig. 1. The inferred parameters of the exponential correlation function were found to be: $G_0 = (0.33 \pm 0.08) \text{ neV}^2$, $\rho_0 = (28 \pm 3.3) \text{ \AA}$. The total cross section of Eq. (5) with these parameters is shown in Fig. 1.

Fig. 2 shows the macroscopic total and transport cross sections of scattering on inhomogeneities:

$$\Sigma_{tr} = \int (1 - \cos\theta) d\Sigma, \quad (7)$$

where θ is the neutron scattering angle.

The difference between total and transport cross sections is about 5% at the neutron energy of 200 neV and is lower at lower energies, it follows that the scattering by inhomogeneities is almost isotropic in this energy range.

The differential cross section of Eq. (6) with inferred parameters G_0 and ρ_0 was used to perform Monte Carlo simulation of the UCN transport in a cylindrical deuterium crystal aiming to obtain the probability for the UCN to go out of the crystal.

The Monte Carlo program used here is the development of our program used earlier for simulation of transport of very slow neutrons in matter, in particular in cold moderators with their temperature changing during UCN motion in a moderator [32].

The UCN were assumed to be born homogeneously with isotropic angular distribution, the reflection from the side walls of a cylinder was specular, from the top and bottom walls – according to the cosine law. The quantum reflection from the boundary deuterium-vacuum at the exit side of the sample was taken into account according to:

$$R = \left(\frac{v_{0,\perp} - v_{\perp}}{v_{0,\perp} + v_{\perp}} \right)^2, \quad (8)$$

where R is the neutron reflection probability, v_{\perp} is the normal to the boundary component of the neutron velocity in medium, $v_{0,\perp} = (v_{\perp}^2 + v_b^2)^{1/2}$ is the normal to the boundary component of the neutron velocity in vacuum, $v_b = 4.46$ m/s is the calculated boundary velocity of deuterium, the boundary energy $E_b = 104$ neV. Recent measurement [33] of the minimal neutron energy from the solid deuterium source $E_{min} = (99 \pm 7)$ neV, which should correspond to the boundary energy of deuterium, does not contradict to this value.

Two variants for the thickness of the solid deuterium sample were chosen: 15 cm - mean thickness of the solid deuterium source in PSI [20] and 8 cm - typical for the UCN source at the TRIGA reactor of Mainz University [16, 22].

The UCN up-scattering due to phonon annihilation was calculated in two variants: the incoherent model [15] and coherent one [25]. The result of this simulation as the probability for the UCN to go out from the cylindrical solid deuterium crystal are shown in Figs. (3-8).

They illustrate effects of temperature, concentration of para-fraction in deuterium, and inhomogeneity of solid deuterium on the probability for the neutrons to leave the moderator.

Fig. 3 shows the probability for the UCN to go out from perfectly homogeneous solid deuterium cylinder with a thickness of 15 cm at different temperatures, at the concentration of para-fraction $c_p = 0$, the up-scattering on phonons was calculated according to the incoherent model of Ref. [15] and the coherent model of Ref. [25]. The following one-phonon annihilation cross sections were used: for the incoherent variant 0.83 b/v, 2.78 b/v, and 7.9 b/v for temperatures 5, 7, and 9 K, respectively; for the coherent variant 0.2 b/v, 0.87 b/v, and 2.78 b/v for the same temperatures. Further the effect of phonon upscattering on the UCN yield will be discussed only for the variant of calculations in the incoherent model, the calculations according to the coherent model predict much lower effect of the

phonon annihilation on the UCN loss in a moderator. It is seen, that the phonon upscattering effect of heating of an ideal deuterium crystal from 5 to 9 K decreases the UCN yield more than 3 times down to $\sim 6.5\%$ of produced neutrons with an energy 150 neV.

Fig. 4 shows the same as in Fig. 3 (thickness of 15 cm, $c_p = 0$), but for the inhomogeneous solid deuterium with the inferred parameters of the exponential correlation function $G_0 = 0.33 \text{ neV}^2$, $\rho_0 = 28 \text{ \AA}$. The effect of inhomogeneities is not very essential for this rather good crystal: neutron yield is decreased 1.3 times and 1.2 times at temperatures 5 and 9 K, respectively, compared to an ideal crystal for the same UCN energy.

The effect of spin-flip upscattering due to admixture of para-deuterium in an inhomogeneous crystal with a thickness of 15 cm at 9 K is seen in Fig. 5. Compared to the effect of phonon annihilation, additional influence of the UCN upscattering with para-ortho transition of deuterium molecules is not as dangerous: at 2% of para-deuterium the UCN yield decreases 22% for neutrons at 150 neV.

Naturally, all the neutron loss effects are larger for thicker sample. Figures 6, 7, and 8 show the neutron energy dependence of the effects of temperature, concentration of para-fraction in deuterium, and inhomogeneity of the crystal on the probability for the neutrons to leave the deuterium moderator with a thickness of 8 cm.

Fig. 6 shows the probability for the UCN to go out from a perfectly homogeneous solid deuterium cylinder with a thickness of 8 cm at different temperatures, at the concentration of para-fraction $c_p = 0$. In this case heating of an ideal deuterium crystal from 5 to 9 K decreases the UCN yield due to the phonon-upscattering effect the same 3 times, but the absolute values of the probability for the neutrons at 150 neV to leave the moderator are larger: 0.36 and 0.12 compared to 0.21 and 0.065 at these temperatures at a thickness of 15 cm.

Fig. 7 shows the same as in Fig. 4 (inhomogeneous solid deuterium, $c_p = 0$), but for a thickness of 8 cm. The effect of inhomogeneities is not essential in this case too: neutron yield is decreased 1.24 times and 1.2 times at temperatures 5 and 9 K, respectively, compared to an ideal crystal for the same UCN energy.

And, at last, Fig. 8 shows the effect of spin-flip upscattering due to admixture of para-deuterium in inhomogeneous crystal with a thickness of 8 cm at 9 K. Compared to the phonon annihilation additional UCN upscattering with para-ortho transition of the deuterium molecules is not as dangerous: at a concentration of para-fraction 2% the UCN yield decreases approximately the same 20% for neutrons at 150 neV.

Maximum UCN losses are seen if we compare, for example, the probability for

the neutrons to leave the sample at the ideal condition of homogeneous material at $T=0$ and in the worst shown variant: inhomogeneous solid deuterium at $T=9$ K, concentration of para-fraction $c_p = 0.02$. In this case for the UCN at 150 neV the yield is 6.5 times lower than from the ideal crystal with a thickness of 15 cm and is only 4.5% of produced neutrons. At temperature of 5 K the UCN yield from a homogeneous crystal is 4.2-4.7 times larger than from a inhomogeneous one at 9 K depending on the para-fraction concentration. For deuterium thickness 8 cm maximum overall losses are lower than for 15 cm: the yield is about 8% of produced neutrons at 9 K and 2% concentration of para-deuterium molecules.

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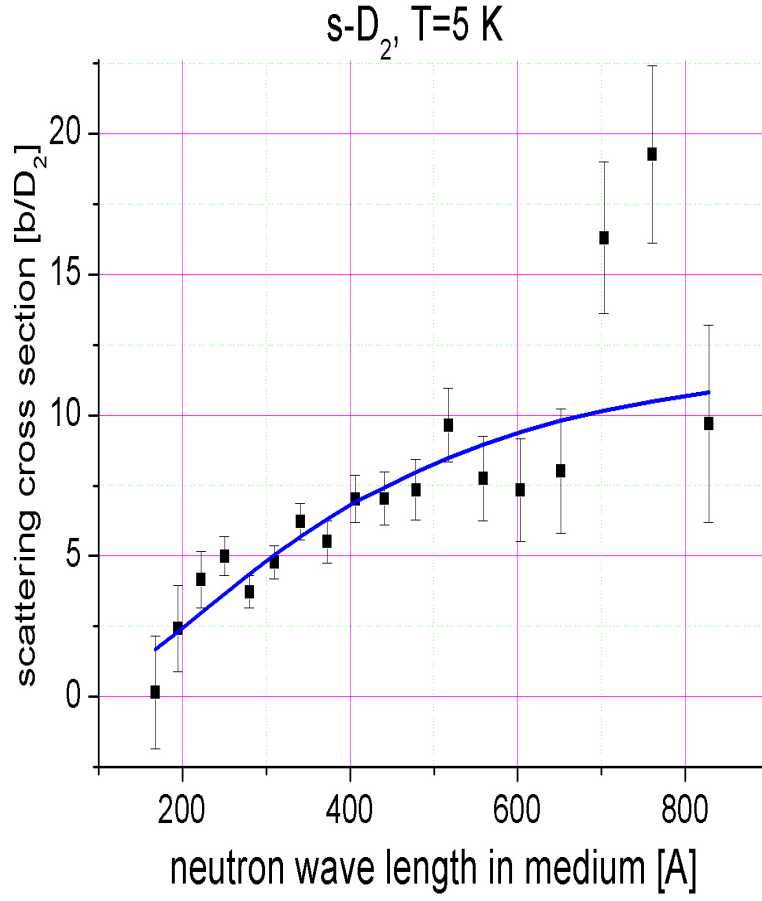


Figure 1: Cross section of scattering on inhomogeneities for the deuterium sample at 5 K (Fig. 2 of [29]) as a function of the in-medium neutron wave length after subtraction of the neutron capture, up-scattering, and the elastic incoherent scattering by deuterium nuclei. The curve is the approximation according to Eq. (5) with the deduced parameters $G_0=0.33 \text{ neV}^2$, $\rho_0=28 \text{ Å}$.

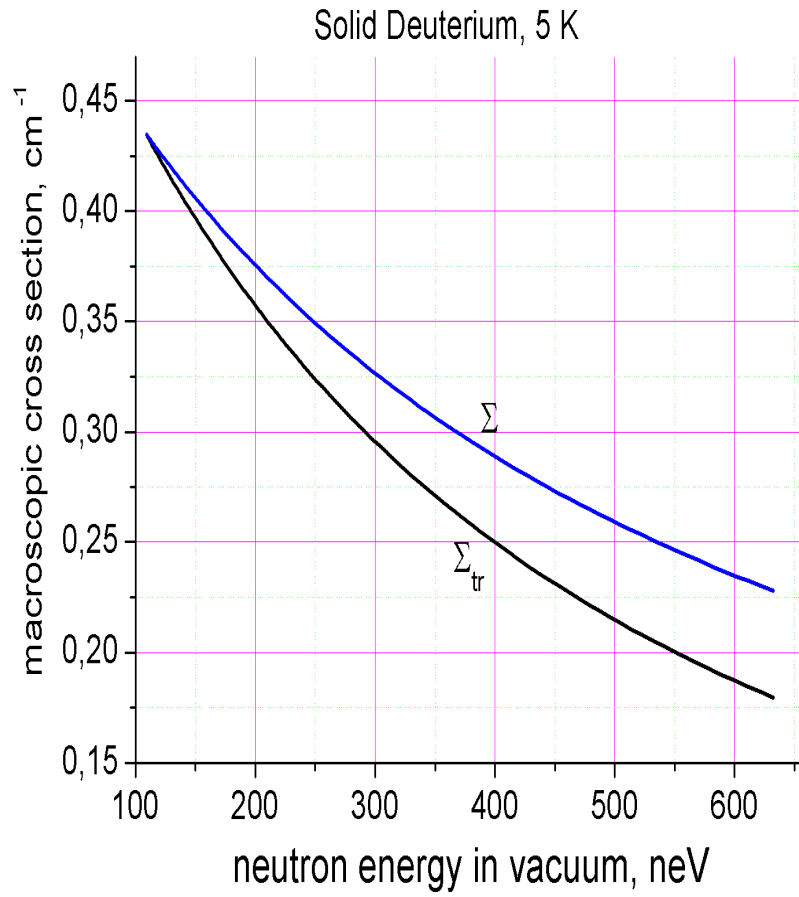


Figure 2: Macroscopic total and transport cross sections of neutron scattering by the solid deuterium crystal inhomogeneities according to Eqs. (5) and (6) with the deduced parameters $G_0=0.33 \text{ neV}^2$, $\rho_0=28 \text{ \AA}$.

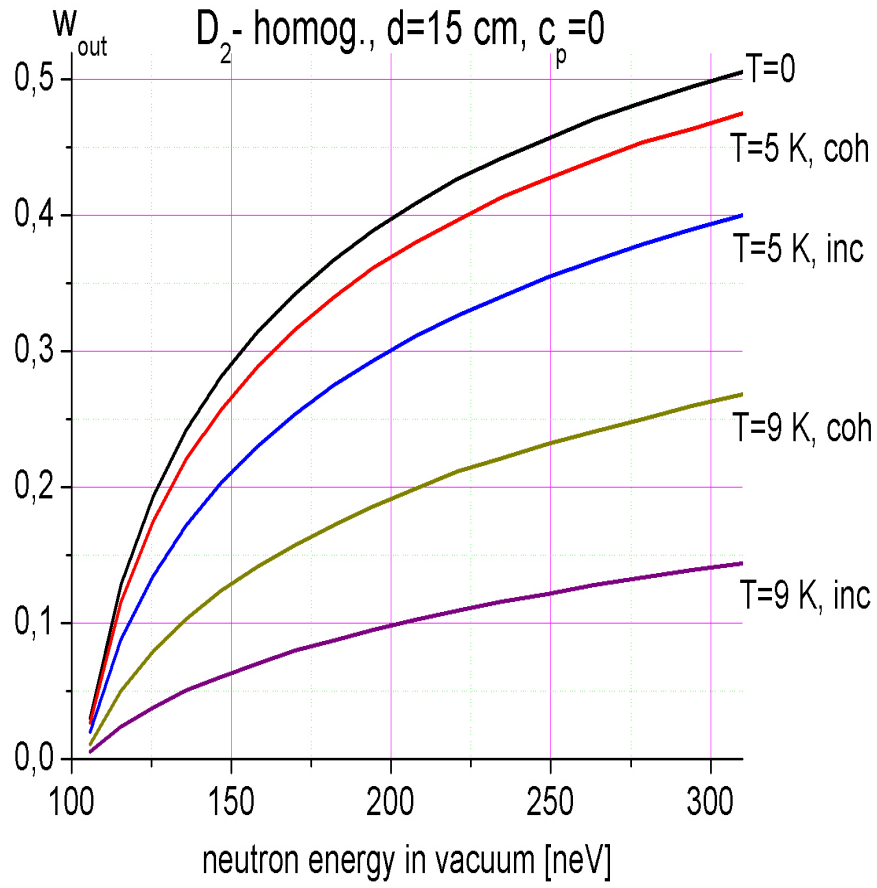


Figure 3: The probability for the UCN to go out from the homogeneous solid deuterium cylinder with thickness 15 cm, the concentration of para-fraction $c_p = 0$, the up-scattering on phonons was calculated according to the coherent model of Ref. [25] and the incoherent model of Ref. [15].

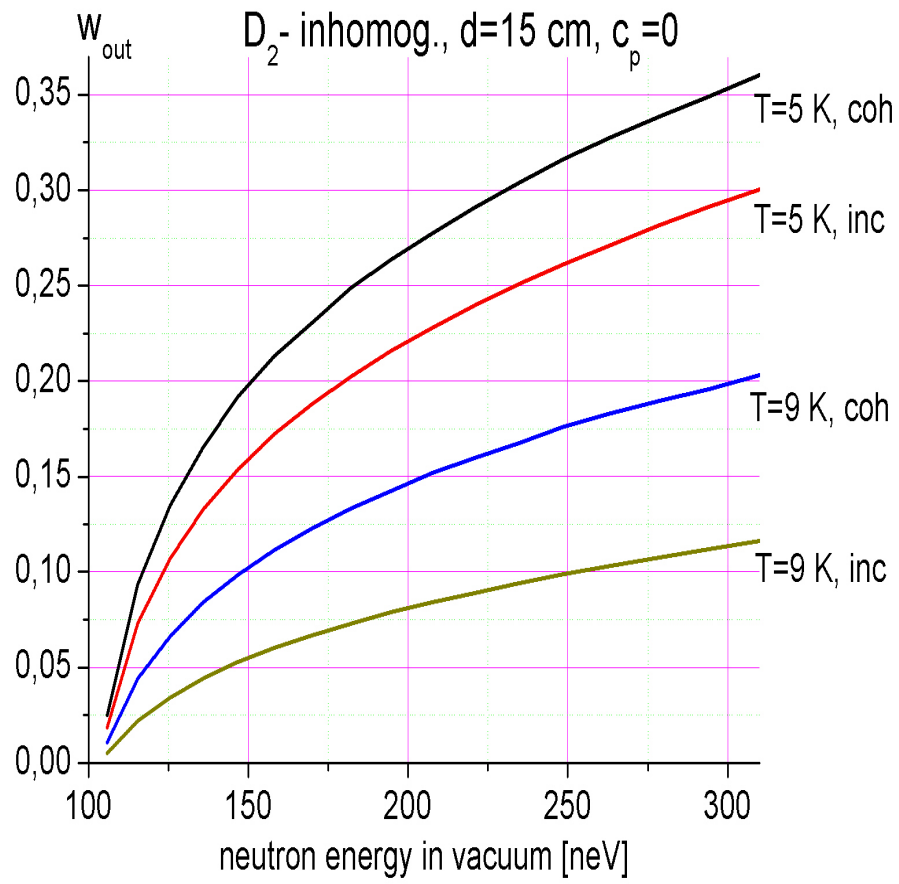


Figure 4: The same as in Fig. 3, but for the inhomogeneous solid deuterium with the inferred parameters of exponential correlation function $G_0 = 0.33 \text{ neV}^2$, $\rho_0 = 28 \text{ \AA}$

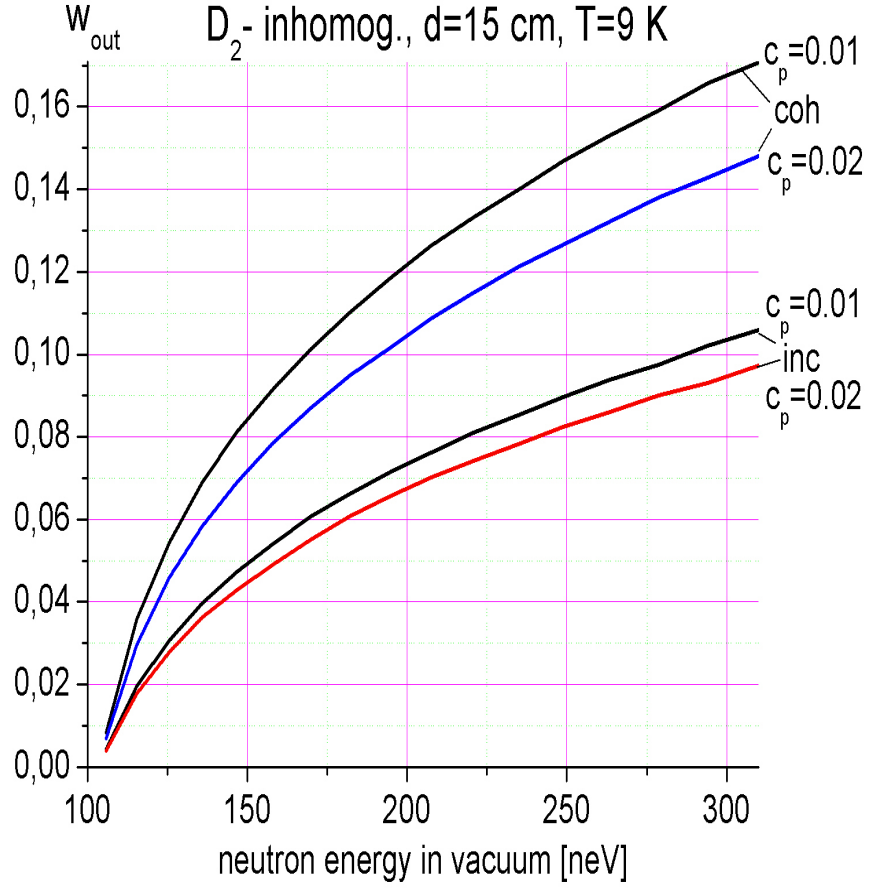


Figure 5: The probability for the UCN to go out from the inhomogeneous solid deuterium crystal with thickness 15 cm and temperature $T=9$ K, at two concentrations of para-fraction $c_p = 0.01$ and 0.02, the up-scattering on phonons was calculated according to the coherent model of Ref. [25] and the incoherent model of Ref. [15].

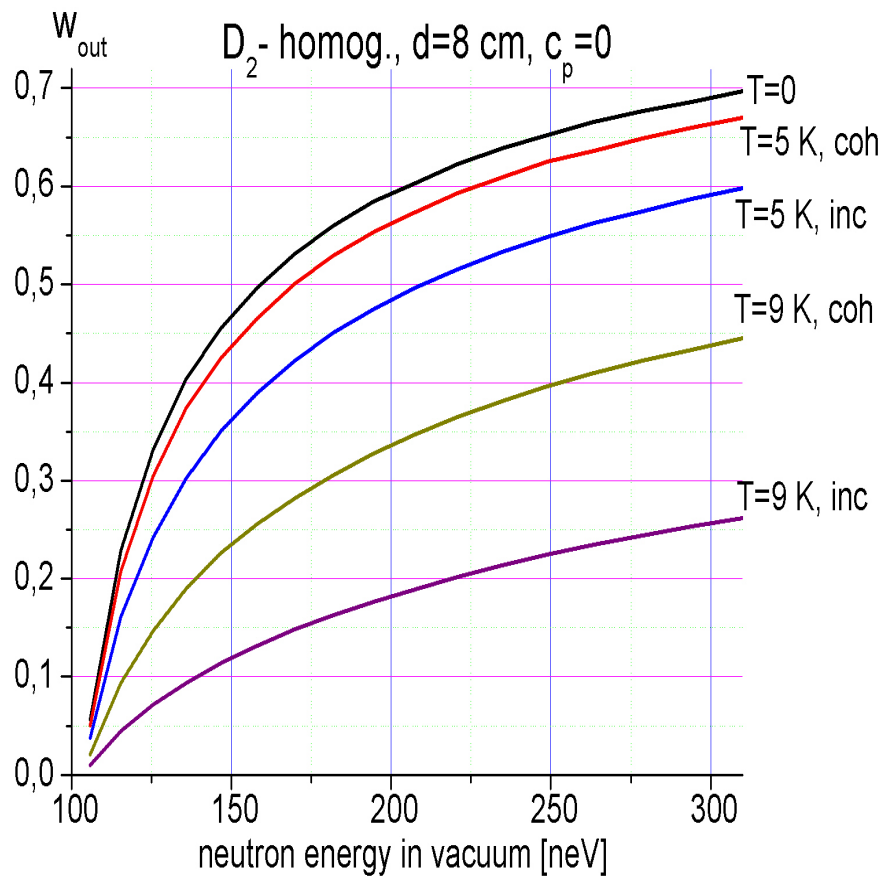


Figure 6: The same as in Fig. 3, but for the deuterium thickness 8 cm.

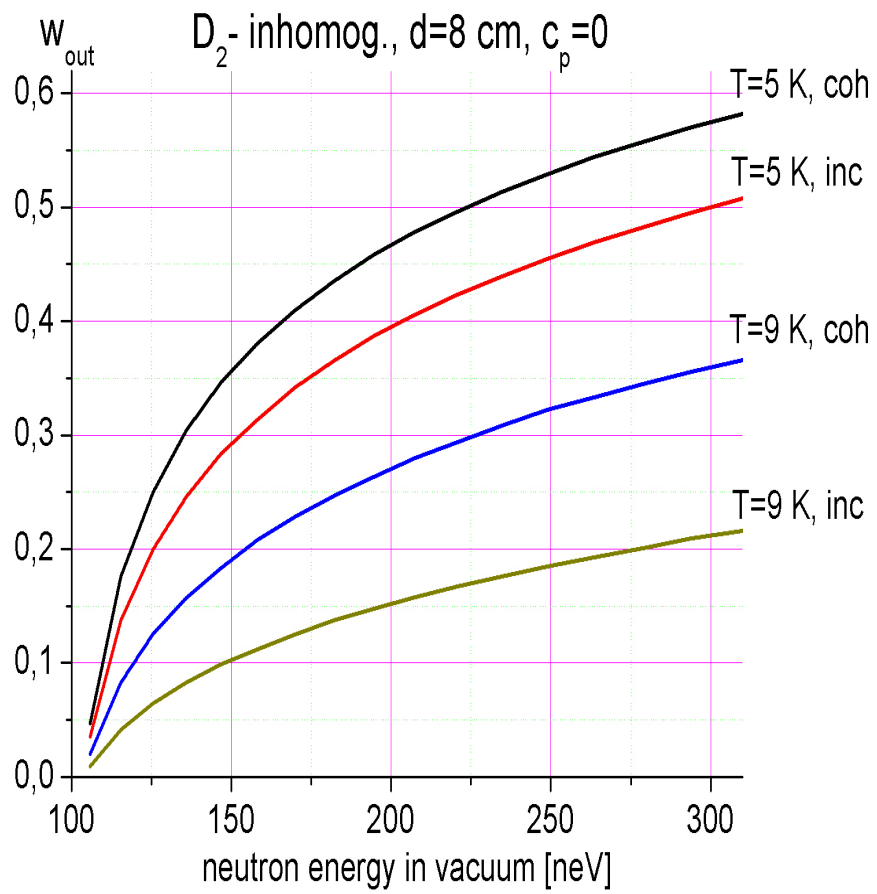


Figure 7: The same as in Fig. 4, but for the deuterium thickness 8 cm.

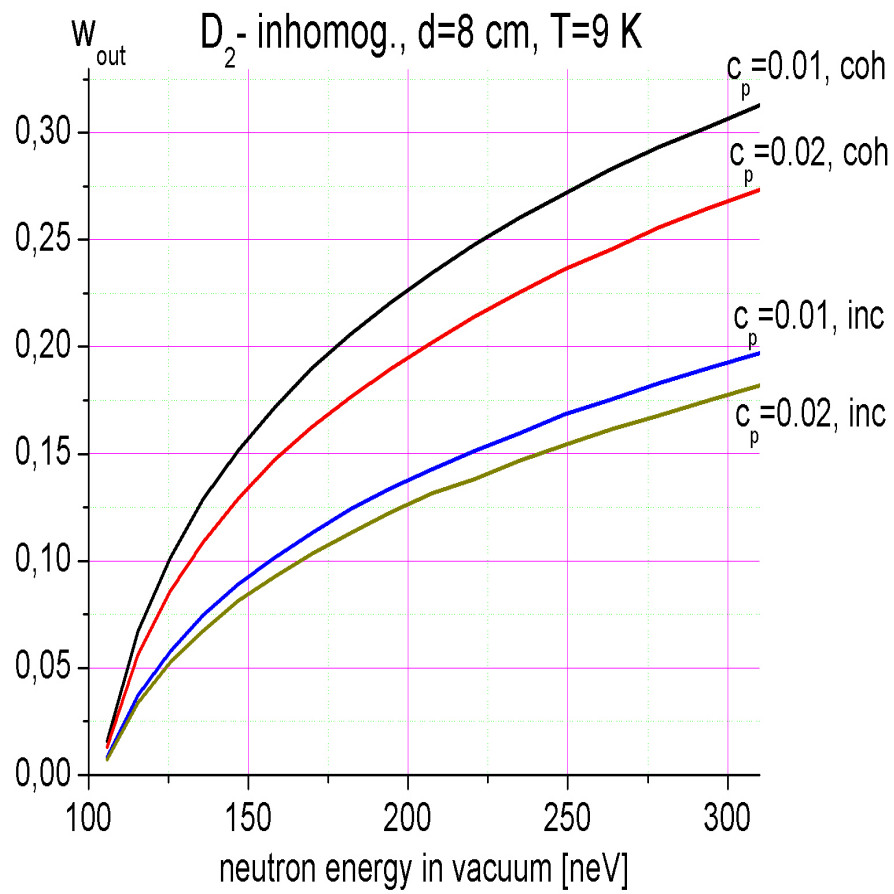


Figure 8: The same as in Fig. 5, but for the deuterium thickness 8 cm.